



UNITED STATES PATENT AND TRADEMARK OFFICE

Commissioner for Patents  
United States Patent and Trademark Office  
P.O. Box 1450  
Alexandria, VA 22313-1450  
[www.uspto.gov](http://www.uspto.gov)

# Fax Cover Sheet

**Date:** 12 Jun 2003

<b>To:</b> Mr. Barry Bretschneider	<b>From:</b> John P. Sheehan
<b>Application/Control Number:</b> 09/987,064	<b>Art Unit:</b> 1742
<b>Fax No.:</b> 202 785 7515	<b>Phone No.:</b> (703) 308-3861
<b>Voice No.:</b> 703-760-7700	<b>Return Fax No.:</b> (703) 872-9310
<b>Re:</b> Translation of Japanese Patent Document	<b>CC:</b>
<input type="checkbox"/> <b>Urgent</b> <input type="checkbox"/> <b>For Review</b> <input type="checkbox"/> <b>For Comment</b> <input type="checkbox"/> <b>For Reply</b> <input type="checkbox"/> <b>Per Your Request</b>	

**Comments:**

Attached is an English language translation of Japanese Patent No. 08-188803 applied in the Office action mailed June 4, 2003.

**Number of pages** 9 **including this page**

## STATEMENT OF CONFIDENTIALITY

This facsimile transmission is an Official U.S. Government document which may contain information which is privileged and confidential. It is intended only for use of the recipient named above. If you are not the intended recipient, any dissemination, distribution or copying of this document is strictly prohibited. If this document is received in error, you are requested to immediately notify the sender at the above indicated telephone number and return the entire document in an envelope addressed to:

Commissioner for Patents  
P.O. Box 1450  
Alexandria VA 22313-1450

PTO 03-3768

CY=JA DATE=19960723 KIND=A  
PN=08-188803

09/987064

PRODUCTION OF POWDERY RARE EARTH-TRANSITION METAL ALLOY  
[Kidorui-senikinzoku gokin funmatsuno seizohoho]

Shotaro Koga

UNITED STATES PATENT AND TRADEMARK OFFICE  
Washington, D.C. June 2003

Translated by: FLS, Inc.

PUBLICATION COUNTRY	(10):	JA
DOCUMENT NUMBER	(11):	08-188803
DOCUMENT KIND	(12):	A
	(13):	PUBLISHED UNEXAMINED PATENT APPLICATION (Kokai)
PUBLICATION DATE	(43):	19960723 [WITHOUT GRANT]
PUBLICATION DATE	(45):	[WITH GRANT]
APPLICATION NUMBER	(21):	06-339216
APPLICATION DATE	(22):	19941229
PRIORITY DATE	(32):	
ADDITION TO	(61):	
INTERNATIONAL CLASSIFICATION	(51):	B 22 F 9/222; H 01 F 1/06
DOMESTIC CLASSIFICATION	(52):	
PRIORITY COUNTRY	(33):	
PRIORITY NUMBER	(31):	
PRIORITY DATE	(32):	
INVENTOR	(72):	KOGA, SHOTARO
APPLICANT	(71):	NOK K.K.
TITLE	(54):	PRODUCTION OF POWDERY RARE EARTH-TRANSITION METAL ALLOY
FOREIGN TITLE	[54A]:	Kidorui-senikinzoku gokin funmatsuno seizohoho

[Claims]

[Claim 1] Production of powdery rare earth-transition metal alloy with the following characteristic:

Coprecipitated fine particles, obtained by coprecipitating a solution consisting of a rare earth element and transition metal salt with an alkali solution, are heated to approx. 600 - 1000°C in air to produce a fine-powdery rare earth element-transition metal composite oxide, which is further heated to 700 to 1,000°C in the presence of a reducing agent under an inert gas for reduction.

[Detailed Explanation of this Invention]

[0001] [Industrial Field]

This invention pertains to a production method of powdery rare earth-transition metal alloy and is particularly associated with a production of powdery rare earth-transition metal alloy having a fixed alloy composition ratio.

[0002] [Conventional Technology]

Rare earth-transitional metal alloy (e.g.,  $\text{SmCO}_5$ ,  $\text{SmCO}_{17}$ ,  $\text{Nd}_2\text{Fe}_{14}\text{B}$ ), being utilized as a permanent magnetic material applied in a wide range of fields, such as electric apparatus motors, sound equipments, and generator, is much researched for developing new materials and improved characteristics.

[0003] Also,  $\text{SmFe}_2$ ,  $\text{TbFe}_2$ , and  $\text{TbDyFe}_2$ , which are a kind of rare earth-transitional metal alloys and magnetic distortion materials producing significant distortion with a magnetic field, are being

researched and developed for the sensor and actuator application fields.

[0004] To produce a rare earth-transition metal alloy utilized in various fields, an alloy formation method utilizing a solubilization technique consisting of the following series of procedures is performed ("Magnetic Magnet Materials for Engineers", pp152, Nikkan Kogyo Shinbunsha, 1991): Raw material composition - Melting - Molding - Rough pulverization - Fine pulverization - molding in magnetic field - sintering - solution annealing - timed treatment - polishing process - product

[0005] However, this method has the following problems:

(1) Due to impure materials mixed during the pulverization process, deterioration of magnetic characteristic is unavoidable.

(2) As the heat processing temperature is so high to cause evaporation of rare earth substances, such as Sm and Dy, controlling the composition is difficult.

(3) As a heat-proof material is required for a high temperature heat processing, production cost cannot be lowered.

(4) Segregation of alloy caused by a peritectic reaction unique to the solubilization method results in composition fluctuation, making it difficult to improve the reproducibility of magnetic characteristic.

[0006] [Problems to Be Solved by this Invention]

The purpose of this invention is to provide a production method of rare earth - transition metal alloy powder that can control the alloy composition without utilizing the conventional solubilization method.

[0007] [Method to Solve the Problems]

The purpose of this invention described above can be achieved by providing a production method of powdery rare earth-transition metal alloy with the following characteristic:

Coprecipitated fine particles, obtained by coprecipitating a solution consisting of a rare earth element and transition metal salt with an alkali solution, are heated to approx. 600 - 1000°C in air to produce a fine-powdery rare earth element-transition metal composite oxide; this powdery composite oxide is further heated to 700 to 1,000°C in the presence of a reducing agent under an inert gas to produce the objective fine-powdery rare earth element-transition metal alloy.

[0008] As a rare earth salt, water-soluble halide or nitrate of a rare earth element such as Dy, Sm, Tb, Ho, is used. Also, as a transition metal element, a water-soluble halide, nitrate, sulfate, oxalate, or the like of a transition metal such as Fe, Co, Ni, is used. Those water-soluble salts are mixed at a specific mol ratio of preparing alloy composition and used as a solution.

[0008] This mixed solution is coprecipitated by adding an alkali solution (e.g., aqueous ammonia, an NaOH aqueous solution, potassium hydroxide solution). In practical application, a mixed solution is dripped in a stirred alkali solution of desired alkali pH; by continuously stirring the solution after the completion of dripping, a suspension liquid is formed. The solution is water-washed until the pH of this suspension liquid reaches 8 - 10. Then, the solution is subjected to filtration. The collected coprecipitated fine particles whose average particle diameter is 0.1 to 1  $\mu\text{m}$  are dried at approx. 20 - 300°C under reduced pressure.

[0010] The acquired dried coprecipitated fine particles are further heated to approx. 600 to 1,000°C, preferably approx. 800 to 1,000°C for approx. 1 - 3 hours in the air to produce a rare earth and transition metal composite oxide powder.

[0011] The produced composite oxide powder is reduced in the presence of a reducing agent such as Ca, Mg, or  $\text{CaH}_2$ , under an inert gas such as Argon or nitrogen, at approx. 700 to 1,000°C, preferably approx. 750 to 850°C for approx. 1/4 - 5 hours. After water washing was repeated to remove the reducing agent included in the reduced product, the objective fine-powdery rare earth element-transition metal alloy can be produced by drying the material at room temperature under reduced pressure.

[0012] [Effectiveness of this Invention]

This invention provides the following effectiveness:

(1) Since a pulverization process required by a solubilization method is not necessary, mixing of impure substances can be avoided, thus preventing the deterioration of magnetic characteristic.

(2) Since the heat-processing temperature is relatively low, controlling the alloy composition is relatively easy, while the production cost can be reduced.

(3) Materials can be made into an alloy in a solid phase reaction without causing the segregation of alloy due to peritectic reaction unique to the solubilization method. Particularly, as an alloy can be prepared using an oxide composite having uniformly dispersed rare earth and transition metals, composition variations in the alloy can be drastically suppressed, thus being able to provide reproducibility of alloy of selected composition ratio.

[0013] [Operational Examples]

The following explains the operational examples of this invention.

[0014] Operational Example:

10.72 g of  $\text{DyCl}_3 \cdot 6\text{H}_2\text{O}$  (purity: 99.9%) and 15.38 g of  $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$  (purity: 99.9%) were dissolved in 100 ml of water to prepare a water solution containing dysprosium and iron water.



[0015] While stirring the ammonia water in a separable flask equipped with a baffle (volume = 500 ml), said water solution was dripped at a speed of 3 ml/min. Then, the mixture was continuously stirred for the next 1 hour. The obtained suspended liquid was washed with water until pH became 8 - 10 and filtered. The collected coprecipitated fine particles were dried at 250°C at reduced pressure, heated at 100°C in the air for 1 hour. As a result, dysprosium and iron oxide composite powder was obtained.

[0016] This oxide composite powder (0.237 g) and calcium hydroxide  $\text{CaH}_2$  (0.684 g) were mixed and heated for 15 minutes at 800°C in argon gas for reduction. Then, after the reduced product was repeatedly washed with water in order to remove the Ca and CaO in the product, the product was dried at room temperature at reduced pressure to prepare dysprosium and iron alloy powder.

[0017] The obtained alloy powder had 5 - 10  $\mu\text{m}$  particle diameter. The atomic composition ratio of this powder was Fe : Dy = 2 : 1 (measured using an induction bonding type plasma emission analysis device). Therefore, the alloy was confirmed as a  $\text{DyFe}_2$  alloy. The measured magnetic characteristic (magnetic distortion constant  $\lambda$ ) of  $\text{DyFe}_2$  prepared from this alloy powder was " $\lambda = 433 \times 10^{-6}$ " with excellent reproducibility.